

Atomistic spin dynamics of low-dimensional magnets

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(Dated: November 14, 2012)

Abstract

We investigate the magnetic properties of a range of low-dimensional ferromagnets using a combination of first-principles calculations and atomistic spin dynamics simulations. This approach allows us to evaluate the ground state and finite temperature properties of experimentally well characterized systems such as Co/Cu(111), Co/Cu(001), Fe/Cu(001) and Fe/W(110), for different thicknesses of the magnetic layer. We compare our calculated spin wave spectra with experimental data available in the literature, and find a good quantitative agreement. We also predict magnon spectra for systems for which no experimental data exist at the moment, and estimate the role of temperature effects.

Keywords: Atomistic spin dynamics; monolayer magnetism; ultrathin magnets; magnons; spin wave; iron; tungsten; cobalt; copper

In recent years, spin-polarized electron energy loss spectroscopy (SPEELS) has been developed to the point of becoming a powerful method for investigating spin waves at surfaces and in thin films.¹ Landmark experiments have included the measurement of the magnon spectrum of ultrathin Co films on Cu(001),² and of a single monolayer (ML) of Fe on W(110).³ More recently, an asymmetry in the magnon spectrum of 2 ML Fe on W(110) has also been reported⁴ - this is a direct signature of the Dzyaloshinskii-Moriya interaction present in this system.⁵ The experimental accessibility of these properties provides an opportunity for comparing the merits of different theoretical approaches used to describe thin-film magnets.⁶⁻⁸ Furthermore, the experiments challenge theoreticians to address issues such as accurately characterizing exotic ground states arising from relativistic spin-orbit coupling effects,^{5,9,10} and treating finite-temperature effects, to name only two.

Previous theoretical studies regarding the spin-wave dispersion in thin layers have been performed by Costa et al.^{7,11}. The method used to investigate these systems consists of an empirical tight binding description, either from empirical data or from first principles calculations, of both the magnetic layers and the substrate followed by a dynamical theory of the spin-waves.¹²⁻¹⁴ Recently the formalism of atomistic spin-dynamics (ASD) was developed¹⁵ and was subsequently implemented in the UppASD package^{16,17}. With this technique it is possible to model spin waves at finite temperature, as shown recently for 1 ML of Fe on W(110).⁸ The method relies on an accurate estimation of the exchange parameters in the Hamiltonian using first-principles calculations, and subsequently evolving it in time according to a modified Landau-Lifshitz-Gilbert equation of motion,^{16,18} as described in the next section.

In this article, we extend the scope of our previous investigations, and employ the first-principles/ASD strategy to calculate the magnon spectra of Co/Cu(001), Co/Cu(111), Fe/Cu(001) and Fe/W(110), for varying thicknesses of the magnetic layer. The chosen systems have been extensively studied experimentally and recently, using SPEELS, also magnon spectra have been measured for selected systems with certain thicknesses of the magnetic overlayer.

I. THEORETICAL APPROACH

First Principles Calculations and Atomistic Spin Dynamics Simulations

Our strategy is to combine first-principles calculations with ASD simulations. In practice, we map an itinerant electron system onto an effective Hamiltonian with classical spins,

$$\mathcal{H} = \mathcal{H}_{\text{ex}} + \mathcal{H}_{\text{SO}}.$$

This generalized Hamiltonian consists of an exchange term:

$$\mathcal{H}_{\text{ex}} = - \sum_{i \neq j} J_{ij} \mathbf{m}_i \cdot \mathbf{m}_j, \quad (1)$$

where i and j are atomic indices, \mathbf{m}_i is the classical atomic moment and J_{ij} is the strength of the exchange interaction, and a spin-orbit interaction term:

$$\mathcal{H}_{\text{SO}} = K \sum_i (\mathbf{m}_i \cdot \mathbf{e}_K)^2 - \sum_{i \neq j} \mathbf{D}_{ij} (\mathbf{m}_i \times \mathbf{m}_j), \quad (2)$$

where K is the strength of the anisotropy field along the direction \mathbf{e}_K and \mathbf{D}_{ij} is the Dzyaloshinskii-Moriya (DM) vector.^{19,20} These represent the main relativistic effects present in low-dimensional magnets, although the magnitude of the DM term is often negligibly small.

The ASD simulations rely on an accurate determination of the exchange integrals and spin-orbit coupling present in the generalized Hamiltonian. In order to determine these, we employed two different methods for our first principles calculations. First, we set up a slab with 9-11 layers representing the substrate, having on one side the magnetic over-layer and a large region of vacuum, simulating the surface. The interlayer distances were relaxed according to the forces using the projector augmented wave method (PAW) as implemented in the VASP program^{21,22}. Once the relaxed geometry is established, we employed the SPR-KKR package^{23,24} for calculations of the parameters to be used within the generalized Hamiltonian using the local spin density approximation (LSDA) for the exchange-correlation potential. The basis set consists of s , p , d and f orbitals and all relativistic effects are included by solving the Dirac equation.

The exchange parameters J_{ij} and Dzyaloshinskii-Moriya (DM) vectors \mathbf{D}_{ij} are obtained from the relativistic generalization by Udvardi *et al.*^{6,25} of the real-space method of in-

infinitesimal rotations of Liechtenstein, Katsnelson and Gubanov (LKG),^{26,27}. Here, the full exchange tensor \bar{J}_{ij} is calculated which can be decomposed into its isotropic part J_{ij} and its anti-symmetric part, which corresponds to the DM-vector. The whole procedure relies on an adiabatic approximation in which the slow motion of the spins is decoupled from the fast motion of the itinerant electrons, a situation that is justified at low energy scales and for systems with reasonably large exchange splitting.

Once calculated, the exchange parameters J_{ij} may be Fourier transformed to obtain the so-called adiabatic magnon spectrum. In the simple case of a single layer (corresponding to one atom per cell), the energy of a spin wave with respect to a ferromagnetic ground state is given by

$$E(\mathbf{q}) = \sum_{j \neq 0} J_{0j} [\exp(i\mathbf{q} \cdot \mathbf{R}_{0j}) - 1], \quad (3)$$

where \mathbf{R}_{ij} is the relative position vector connecting sites i and j . From this it is straightforward to calculate the spin wave dispersion $\omega(\mathbf{q})$.²⁸ For systems with more than one atom per cell, as is the case for thin films consisting of more than one monolayer, the spin wave energies are given by the eigenvalues of the general $N \times N$ matrix here expressed in block form

$$\begin{bmatrix} \sum_j^N J_0^{ij} - J^{ii}(\mathbf{q}) & -J^{ij}(\mathbf{q}) \\ -J^{ij}(\mathbf{q})^* & \sum_i^N J_0^{ji} - J^{jj}(\mathbf{q}) \end{bmatrix}, \quad (4)$$

where N is the number of atoms per cell (*i.e.* in this case the number of monolayers).

Using in the ASD simulations the generalized Hamiltonian \mathcal{H} as a starting point, the effective interaction field experienced by each atomic moment \mathbf{m}_i is given by

$$\mathbf{B}_i = -\frac{\partial \mathcal{H}}{\partial \mathbf{m}_i}.$$

The temporal evolution of the atomic spins at finite temperature is governed by Langevin dynamics, through coupled stochastic differential equations of the Landau-Lifshitz form,

$$\frac{\partial \mathbf{m}_i}{\partial t} = -\frac{\gamma}{(1 + \alpha^2)} \mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)] - \gamma \frac{\alpha}{m(1 + \alpha^2)} \mathbf{m}_i \times \{\mathbf{m}_i \times [\mathbf{B}_i + \mathbf{b}_i(t)]\}, \quad (5)$$

where γ is the the electron gyromagnetic ratio. Temperature fluctuations are included via a stochastic Gaussian shaped magnetic field $\mathbf{b}_i(t)$ with properties $\langle \mathbf{b}_i(t) \rangle = 0$ and

$$\langle b_i^k(t)b_j^l(t') \rangle = 2D\delta_{ij}\delta_{kl}\delta(t-t'), \quad D = \frac{\alpha}{(1+\alpha^2)} \frac{k_B T}{\mu_B m}, \quad (6)$$

where i and j denote lattice sites, k and l the cartesian components and α is the Gilbert damping parameter which eventually brings the system to thermal equilibrium.

The coupled equations of motion (5) can be viewed as describing the precession of each spin about an effective interaction field, with complexity arising from the fact that, since all spins are moving, the effective field is not static. In our calculations we evolve the stochastic Landau-Lifshitz equations using a semi-implicit method introduced by Mentink *et al.*¹⁸

The principal advantage of combining first-principles calculations with the ASD approach is that it allows us to address the dynamical properties of spin systems at finite temperatures.^{16,29,30} We focus in particular on two important quantities, the space- and time-displaced correlation function:

$$C^k(\mathbf{r} - \mathbf{r}', t) = \langle m_{\mathbf{r}}^k(t)m_{\mathbf{r}'}^k(0) \rangle - \langle m_{\mathbf{r}}^k(t) \rangle \langle m_{\mathbf{r}'}^k(0) \rangle, \quad (7)$$

where the angular brackets signify an ensemble average and k the cartesian component, and its Fourier Transform, the dynamical structure factor:

$$S^k(\mathbf{q}, \omega) = \frac{1}{\sqrt{2\pi N}} \sum_{\mathbf{r}, \mathbf{r}'} e^{i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}')} \int_{-\infty}^{\infty} e^{i\omega t} C^k(\mathbf{r} - \mathbf{r}', t) dt, \quad (8)$$

where \mathbf{q} and ω are the momentum and energy transfer, respectively. The dynamical structure factor, $S(\mathbf{q}, \omega)$ is the quantity probed in neutron scattering experiments of bulk systems³¹, and can analogously be applied to SPEELS measurements. By plotting the peak positions of the structure factor along particular directions in reciprocal space, the spin wave dispersions may be obtained.^{8,16,29,30}

II. RESULTS

A. Critical temperatures

Within the Heisenberg model and in the case where the anisotropy and finite size effects are absent, a two dimensional magnetic film cannot sustain long range magnetic order at

finite temperatures (the celebrated Mermin -Wagner theorem). However, the presence of symmetry breaking perturbations, such as small anisotropies, will overrule Mermin-Wagner theorem and finite temperature magnetism is possible, even in the case of a single magnetic layer. The critical temperatures are often reduced in thin films compared to their bulk counterpart. Using our *ab initio* calculated exchange interaction parameters, anisotropy and magnetic moments, we estimate the critical temperature, T_c , in several thin magnetic films by means of Monte Carlo simulations and using the cumulant crossing method³². The systems we investigated are $\text{Co}_N/\text{Cu}(111)$, $\text{Co}_N/\text{Cu}(001)$, $\text{Fe}_N/\text{Cu}(001)$ and $\text{Fe}_N/\text{W}(110)$, where N indicates the number of magnetic atomic layers present. The results are summarized in Table I. It is difficult to directly compare the calculated T_c values with experimental results^{33,34}, due to the lack of information about interface quality, inter-diffusion between the substrate and the magnetic film and the surface roughness of the samples. All these imperfections affect the value of the critical temperature.³⁵ The calculations assume the idealized case with perfect interfaces and are likely to give the highest possible value of T_c that the system can reach. Our data for 3 ML Co/Cu(111) are in rather good agreement with the measured values for 2.5 ML Co/Cu(111), although a comparison should be done with care, since the experiment and theory have slightly different Co thickness. For the 1ML Co/Cu(111) system as well as for the bulk Co, the theoretical values seem to be in agreement with measurements, whereas for the 2ML Co/Cu(001) system the calculated value of T_c is two times larger than the measured value. For 2ML Fe/W(110) as well as for bulk Fe, theory and experiments are in good agreement, whereas for the Fe/Cu(001) system, theory consistently overestimates the measured values. Our calculations do however agree reasonably well with the results obtained by Pajda *et. al*³⁶ using the random phase approximation (RPA).

B. Spin wave spectra

1. Co on Cu(001)

The fcc Co/Cu(001) system represents a model system for the study of magnetic phenomena in thin films, since it does not exhibit strong structural, chemical or magnetic instabilities.^{33,42} It was therefore a natural candidate for investigation with SPEELS, and became the first ultrathin system in which its magnon spectrum was measured using this method.² The system could be described to a good level of accuracy within the context of the nearest-neighbour Heisenberg model on a semi-infinite substrate. In this case a surface

Table I. Calculated values of the Curie temperatures T_C for Co and Fe overlayers on W and Cu substrates with different orientations and compared with published values determined theoretically and experimentally.

System	T_C (K)		Experiment
	Theory: this work	previous studies	
bulk fcc Co	1120	1080 ³⁷ , 1250 ³⁸ , 1311 ± 4 ³⁹	1388-1398
<i>Co_N/Cu111</i>			
1	255	—	207-434 ³³
2	660	—	500 [1.7 ML] ³³
<i>Co_N/Cu001</i>			
1	370	426 ³⁶	—
2	680	—	325 ³³
3	705	—	650 [2.5 ML] ⁴⁰
8	1070	—	—
<i>Fe_N/Cu001</i>			
1	400	515 ³⁶	≈ 100 ³³
2	380	—	≈ 270 ³³
3	560	—	≈ 290 ³³
<i>Fe_N/W110</i>			
2	520	—	450 ⁴¹

mode exists with a dispersion curve $\hbar\omega^{(\text{surf})} = 8JS(1 - \cos(qa_0))$ along the $\langle 110 \rangle$ direction, where J is the exchange coupling, S is the magnitude of the spin per primitive unit cell, q is the length of the magnon wave vector and $a_0 = 2.55 \text{ \AA}$. The fit of this curve to the measured SPEELS data gave $JS = 15.0 \pm 0.1 \text{ meV}$, which compares well with the value of $JS = 14.7 \pm 1.5 \text{ meV}$ obtained from neutron data of bulk fcc Co at long wave lengths⁴³ (*i.e.* in the regime $q < 0.3 \text{ \AA}^{-1}$).

The Halle group followed up their initial report for the 8 ML film² with results relevant to Co/Cu(001) systems with decreasing thickness down to 2.5 ML.^{44,45} These experiments demonstrated a very weak reduction in the energies required to excite the spin waves, relative to the bulk.⁴³ However, for all the thicknesses reported, the surface mode at the surface Brillouin zone boundary (the \bar{X} point) is well below the bulk band edge, at around 240 meV. This difference is for the most part caused by the reduced number of nearest neighbours (NN) at the surface (only 8 atoms) with respect to the bulk case, where there are 12 NN present.² For energy ranges where Stoner excitations are important, no direct comparison between calculated and experimental data for bulk Co is possible at the moment.

Theoretical investigations of Co/Cu(001) thin films have been carried out using both the adiabatic approximation^{6,36} and the random phase approximation (RPA) to a description of the spin response of the itinerant electron system.^{12,13} The primary difference between the two approaches concerns the treatment of the particle hole excitations known as Stoner excitations,⁴⁶ which are neglected in the adiabatic case. Broadly speaking, these are not relevant at low energies, where the density of magnon states dominates. However, at higher energies or large wave-vectors their role becomes significant, and one would therefore expect theories based on the adiabatic approach to break down at the Brillouin zone boundary. In addition, Stoner excitations are more pronounced for low dimensional magnets compared to bulk magnets.

Fig. 1(a) displays the calculated adiabatic magnon spectrum obtained for 8 ML Co/Cu(001), using Eq. (4) with J_{ij} values obtained using the LKG method.^{26,27} The most notable feature is the presence of several branches, one for each Co layer present. This is in contrast to experimental observations, where only the lowest ("acoustic") branch² (filled circles in Fig 1(a)) as well as the second lowest branch¹¹ are observed. As Vollmer *et al.* point out,² this indicates the shortcomings of a direct interpretation of their data in terms of a naïve Heisenberg model.

Fig. 1(b) displays the adiabatic magnon spectrum obtained for 3 ML Fe/Cu(001). Unfortunately, there are no experimental data to compare with. The values for the spin wave stiffness, D , can be estimated from the calculated adiabatic magnon spectra by measuring the curvature of the dispersions as $\mathbf{q} \rightarrow 0$. For this system we find D being approximately $210 \text{ meV } \text{\AA}^2$. For the 1 ML Co on Cu(001) case we obtain a value of the order of $420 \text{ meV } \text{\AA}^2$ - a 15 % overestimate of the experimentally determined value of $360 \text{ meV } \text{\AA}^2$, but considerably softer than the theoretical value determined by Pajda *et al.* of 532 ± 9 ,³⁶ obtained by a real-space adiabatic approach. For 1 ML Fe on Cu(001) we obtain a lower value, of $260 \text{ meV } \text{\AA}^2$ compared to the value of $331 \text{ meV } \text{\AA}^2$ by Pajda *et al.*³⁶.

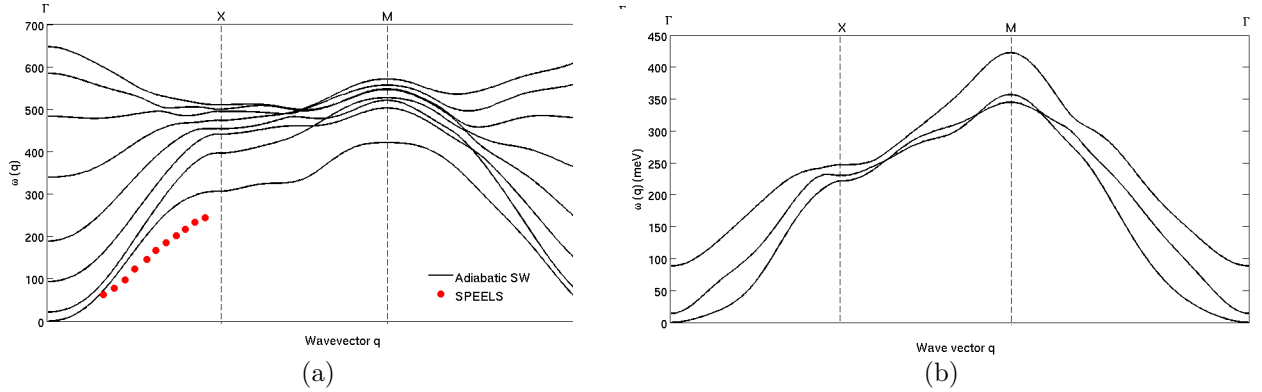


Figure 1. : Adiabatic magnon spectra (full lines) obtained from SPR-KKR calculations for (a) 8 ML Co/Cu(001) together with experimental SPEELS data² (circles) and (b) 3 ML Fe/Cu(001).

As explained above, the adiabatic approximation becomes questionable at higher energies and/or large wave-vectors, since Stoner excitations become relevant. In order to address this issue, Costa *et al.*^{12,13} have developed a theory that explicitly takes these excitations into account. Their approach successfully describes the SPEELS measurements for the 8 ML Co/Cu(001) system: their calculated magnon spectrum is in agreement with the experiment over all the $\bar{\Gamma} - \bar{X}$ line and correctly predicts a broadening of the “acoustic” spin wave peaks, along with an absence of standing spin waves giving rise to “optical” branches. Their work strongly indicates that a process known as Landau damping, through which the spin waves decay into the Stoner continuum, is at play in the Co/Cu(001) system.

At present, the ASD method cannot handle one-particle Stoner excitations directly (these are responsible for the longitudinal fluctuations of the magnetic moments), although recently efforts have been made to include these effects in the LLG-equation, both directly⁴⁷ and

indirectly⁴⁸. However, the ASD method allows to address an alternative source of damping, namely the temperature, and we have performed simulations for different thicknesses of Co overlayers (1, 2, 3 and 8 atomic layers) under two very different conditions (Figs. 2 – 6). First we performed simulations at low temperature (1 K), where the damping constant α in Eq. (5) is set to 3×10^{-4} , a value that ensures a very weak coupling to the temperature bath. In this case the temperature effects are deliberately kept to a minimum. Secondly, we perform simulations in more realistic conditions, namely at room temperature ($T=300$ K) and a physically more plausible damping constant $\alpha = 0.05$. All these results are discussed in detail in the following sections.

1 ML

Fig. 2 displays the spin wave spectra in a 1 ML thick Co layer on Cu(001). In the low temperature case (Fig. 2(a)), the spectra obtained from the atomistic spin dynamics simulations is almost indistinguishable from the adiabatic spectra. We point out that these results are obtained from a calculation of the dynamical structure factor $S(\mathbf{q}, \omega)$. In order to facilitate the comparison between the spin wave spectra for a single layer of Co on Cu for different orientations of the substrate, we performed the calculations for both Cu(001) and Cu(111) orientations (discussed in detailed below), at the same value of the temperature, i.e. $T=200$ K. Increasing the temperature and damping (Fig. 2(b)) causes some broadening of the dynamical structure factor and as a result the magnon energy at the BZ boundary (\bar{X} and \bar{M} points) decreases slightly. However, overall the effects of temperature and dynamic treatment are not significant in this case.

2 and 3 ML

Qualitatively, the 2 ML and 3 ML case are not so different so we only display the calculated dynamic structure factor for 2 ML Co in Figure 3. Having more than one layer on top of Cu(001), we expect, apart from the acoustic branch, a number of optical branches to appear in the magnon spectra, similar to what is found in the adiabatic spectra. In the case of 2 ML we expect one branch of each kind, but as noticed in Fig. 3(a), even at very low temperature ($T=1$ K) and extremely small damping, the optical branch is very weak,

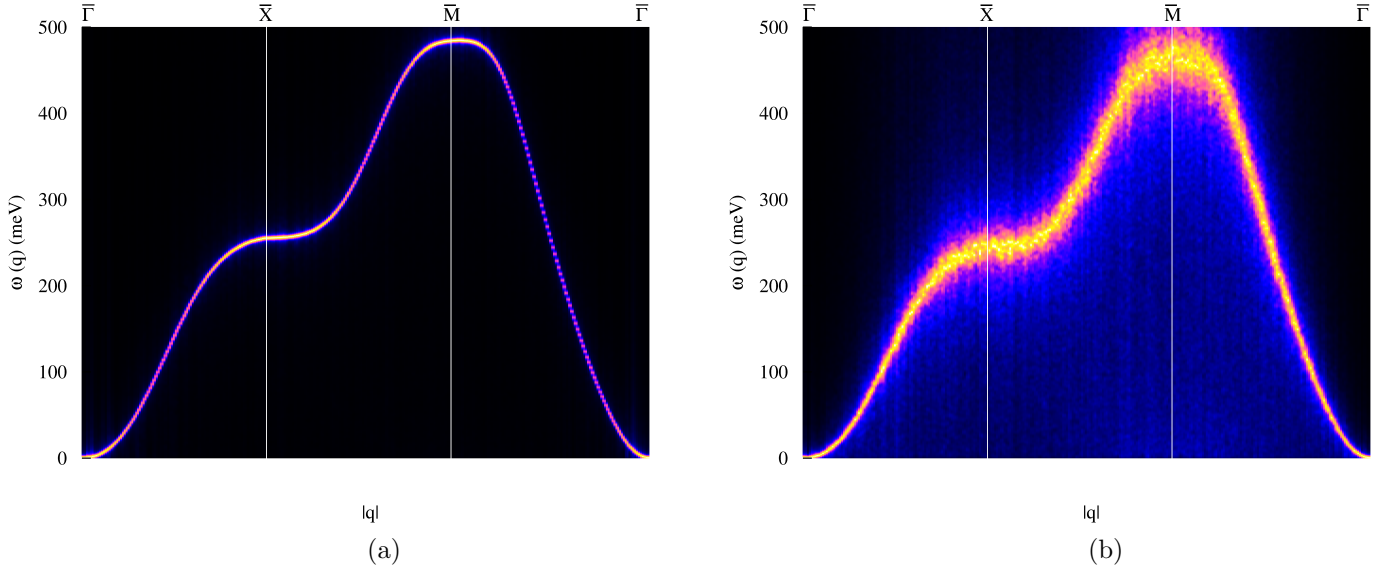


Figure 2. Spin wave dispersion spectra obtained from ASD simulations of 1 ML Co/Cu(001) at (a) $T = 1\text{ K}$ and small damping constant $\alpha = 3 \times 10^{-4}$ and (b) $T = 200\text{ K}$ and realistic damping constant $\alpha = 0.05$

especially at small wave vectors, close to the $\bar{\Gamma}$ -point. In Section II C below, we make a deeper analysis of the dynamical structure factor to better understand the suppression of the optical branches. This type of analysis has been recently published by Taroni et al..⁴⁹ Using a more reasonable temperature and damping (Fig. 3(b)) erases most traces of the optical branch which is smeared out and remains slightly visible only in the $\bar{X} - \bar{M}$ region. The magnon energy at the BZ boundaries is reduced in this case by roughly 25 meV which can be attributed to temperature effects.

8 ML

Fig. 4 displays the spin wave spectra on the full 8 ML Co stack on Cu(001). It is immediately noticed that the contrast between the calculated spectrum based on the dynamical structure factor (Fig. 4(a)) with the spectrum directly obtained from first-principles (Fig. 1(a)) is strong: for values of q close to the $\bar{\Gamma}$ -point. In Fig. 4, there is little trace of the “optical” branches even in the case of low temperature and damping. We also note that at 300 K, the magnon energies at the zone boundaries are reduced by roughly 25 meV, similar to the 2 ML case. The suppression of optical branches arising from a dynamical treatment

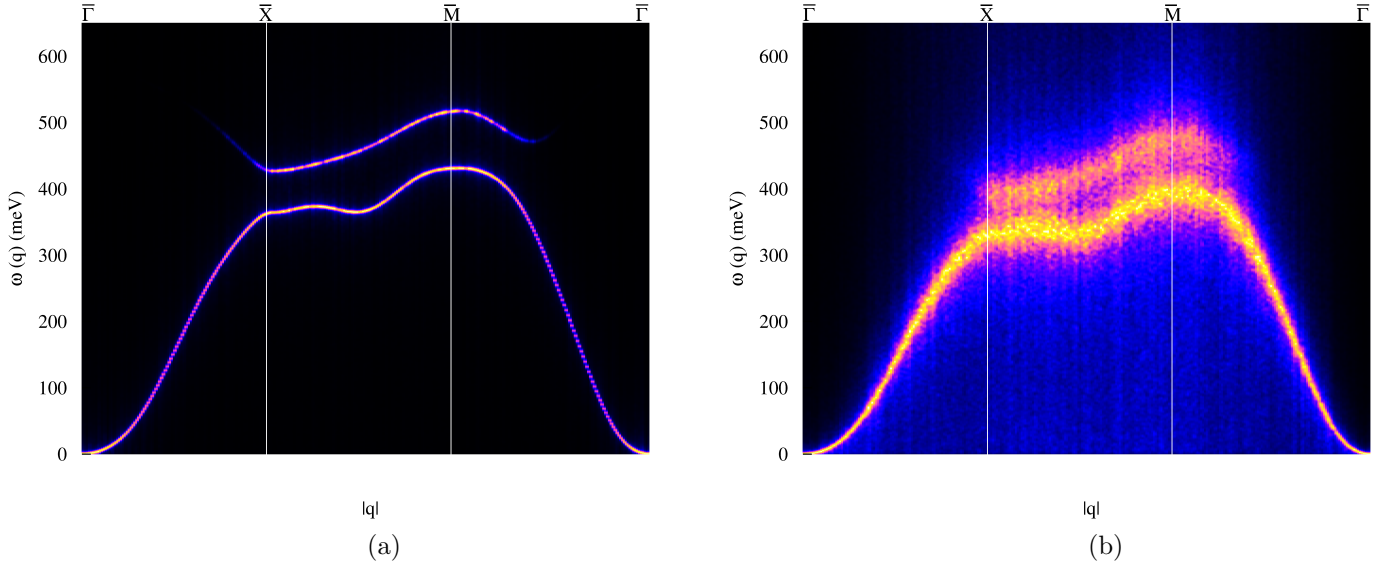


Figure 3. Spin wave dispersion spectra obtained from ASD simulations of 2 ML Co/Cu(001) at a) $T = 1\text{ K}$ and small damping constant $\alpha = 3 \times 10^{-4}$ and b) $T = 300\text{ K}$ and realistic damping constant $\alpha = 0.05$

has also been pointed out by Costa *et al.*^{12,13}. We also note that the agreement between the theoretical acoustic branch that we calculated and experiment is rather good.

2. Co on Cu(111)

For completeness, we also performed simulations on a single layer of Co on the Cu(111) surface. Due to the fact that the critical temperature of the single Co layer on Cu(111) is lower than the room temperature (i.e. $T_c = 255\text{ K}$), for this system we performed the high-temperature calculations at 200 K instead of room temperature. In Fig. 5, the spin wave spectra obtained at $T = 200\text{ K}$ is displayed. The calculated spin wave spectra is noted to be softer than that of the 001-surface, which is consistent with the lower Curie temperatures obtained for this system. The calculated spin wave stiffness has a value of around $220\text{ meV}\text{\AA}^2$, almost half of the value compare to what was found for the 001-surface.

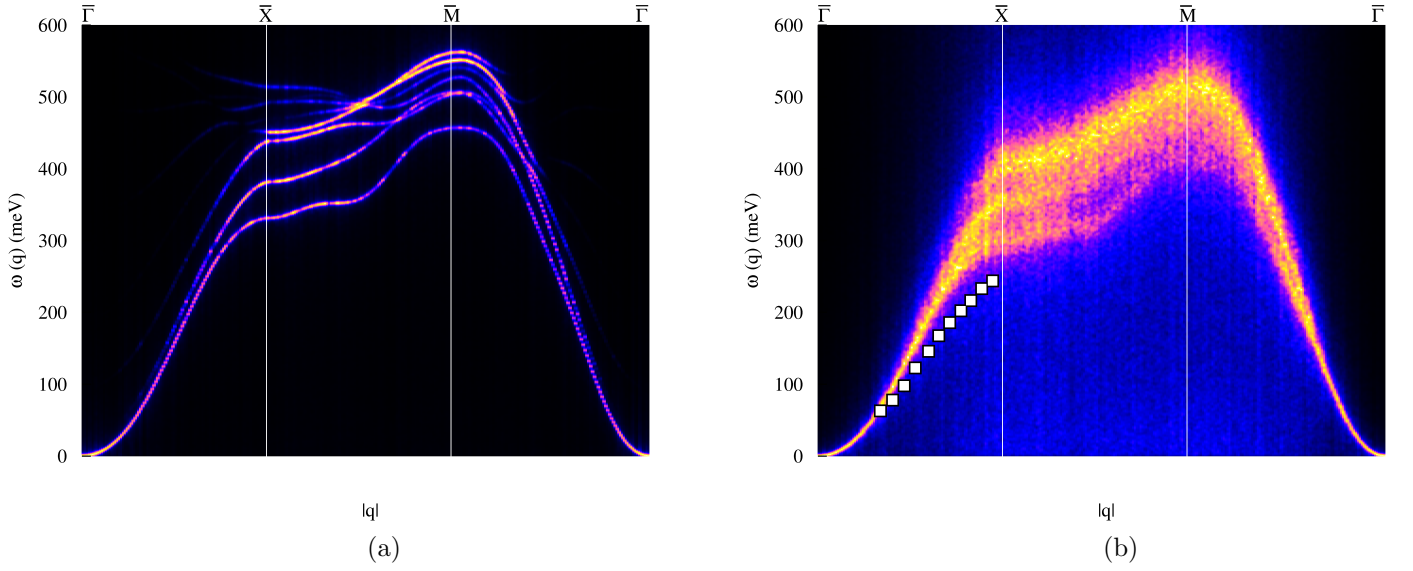


Figure 4. Spin wave dispersion spectra obtained from ASD simulations of 8 ML Co/Cu(001) at a) $T = 1\text{ K}$ and small damping constant $\alpha = 3 \times 10^{-4}$ and b) $T = 300\text{ K}$ and realistic damping constant $\alpha = 0.05$. The experimental values are marked by white squares.²

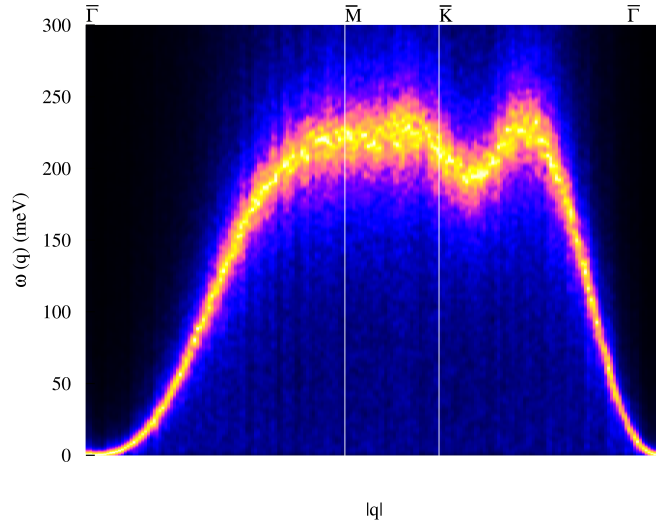


Figure 5. Spin wave dispersion spectra obtained from ASD simulations of 1 ML Co/Cu(111) at $T = 200\text{ K}$ and realistic damping constant $\alpha = 0.05$

3. Fe on Cu(001)

The Fe/Cu(001) system is more complex than Co/Cu(001) for a number of reasons. Since Fe doesn't naturally exist in the fcc phase it is difficult to grow thicker layers of Fe with good

quality. It is also well known, from theory, that bulk fcc Fe exhibits a complicated magnetic phase diagram with many magnetic configurations with similar energies. The same thing is true for thick Fe layers on Cu. However, for thin Fe layers (1 to 3 ML), it is generally accepted that Fe adopts a ferromagnetic configuration. In the case of thicker layers there are several proposed magnetic configurations, for instance Sandratskii⁵⁰ claimed that the magnetic structure takes the form $\downarrow\uparrow\uparrow$ for the three upper layers. We performed Monte Carlo simulations using exchange parameters starting from either ferromagnetic or the proposed magnetic structure of Sandratskii as reference state for the 3 ML case. Regardless of the starting configuration, we always obtained the ferromagnetic configuration as the ground state magnetic structure also for 3 ML Fe on Cu001. However, the spin waves (Fig. 6) are soft which is reflected in the calculated adiabatic spin wave stiffness constant D for which we obtain a value of $170 \text{ meV}\text{\AA}^2$, which is considerably lower than what we obtain for 1 ML. We would like to point out that in general ASD gives a more realistic description of the magnon spectra than the adiabatic approximation (Fig. 1(b)).

In Fig. 6 we show the results of spin wave spectra only for the 3 ML case. The spectra is rather different from Co/Cu(001) and the most striking difference is the overall softness of the magnons. However, we should keep in mind that the critical temperature for the 3 ML Fe/Cu(001) is much lower than for the analogous Co system. The optical branch for the Fe system is more pronounced at low temperatures but similar to the Co case it is suppressed close to the $\bar{\Gamma}$ -point for reciprocal vectors inside the Brillouin zone. For realistic conditions, i.e. at room temperature and for larger damping, the intensity of the optical branches is very weak and smeared traces of those branches remains visible mostly in the $\bar{X} - \bar{M}$ region.

4. Fe on W(110)

In a previous study⁸, we performed calculations of the spin wave spectra of a single Fe layer on top of W(110) and compared it with the experimentally determined spectra³ and found a good agreement. In particular, we observed a softening of the magnons with respect to the bulk spin wave dispersion spectra. This magnon softening in thin layers is expected even when considering a simple NN Heisenberg model for the description of the dispersion curves in ferromagnets, where the exchange parameters are weaker due to a lower coordination at the surface. Here we focus on the magnons of a Fe bilayer on W(110). Experimentally, the

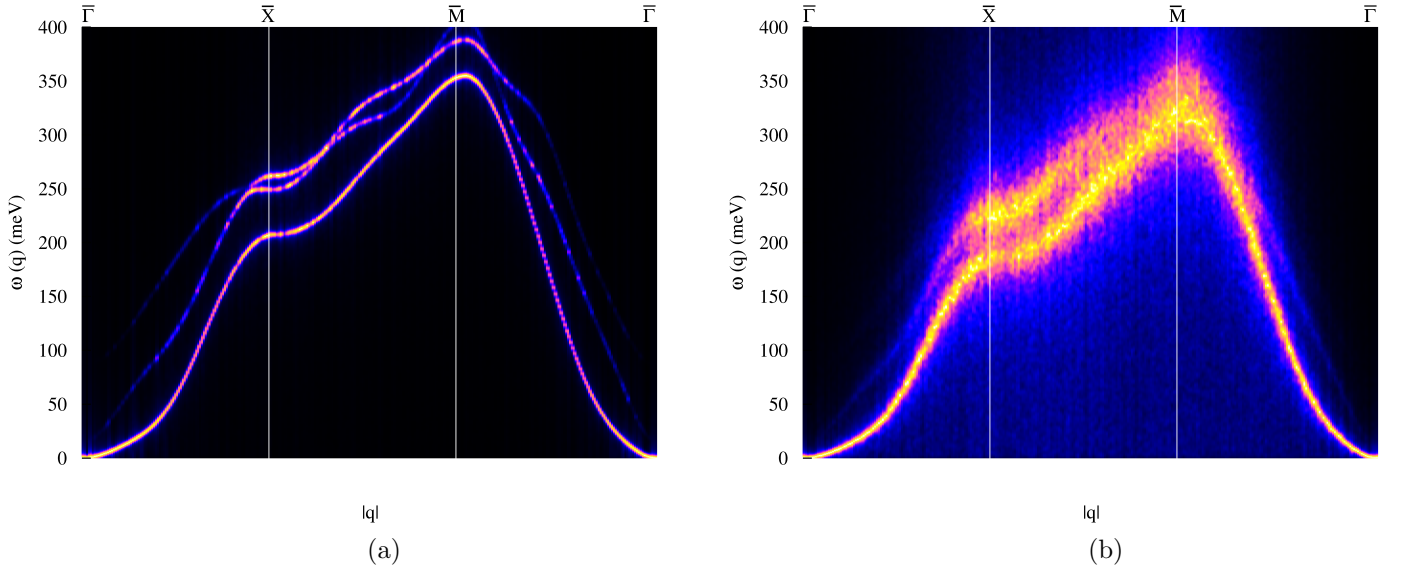


Figure 6. Spin wave dispersion spectra obtained from ASD simulations of 3 ML Fe/Cu(001) at a) $T = 1\text{ K}$ and small damping constant $\alpha = 3 \times 10^{-4}$ and b) $T = 300\text{ K}$ and realistic damping constant $\alpha = 0.05$

magnon dispersion has been measured by SPEELS⁵¹ where in addition the magnon lifetimes were determined⁵². In Fig. 7 we show our calculated magnon dispersion along symmetry lines in the first Brillouin zone together with experimental data. Along the line $\bar{\Gamma} - \bar{H}$, we find a very good agreement between experiment and theory (Fig. 7(a)). If the momentum transfer is changed, so that one goes outside the first Brillouin zone, the different spin wave branches can be determined. This is analyzed in detail in Section II C. In Fig. 7(b) we show the so obtained magnon dispersion, where the optical modes can be clearly observed.

Udvardi and Szunyogh⁵ predicted the asymmetry in the magnon spectrum arising from the Dzyaloshinskii-Moriya interaction. Later on it was experimentally detected by Zakeri *et al.*⁴ in a Fe bilayer on W(110). The spin wave asymmetry ΔE is defined as the difference in the spin wave energy $\omega(\mathbf{q})$ between \mathbf{q} and $-\mathbf{q}$, i.e. $\Delta E = \omega(\mathbf{q}) - \omega(-\mathbf{q})$. If Dzyaloshinskii-Moriya interactions are absent, i.e. very weak effect from spin-orbit interaction, the asymmetry is zero for every wave vector \mathbf{q} . In Fig. 8, we show our calculated spin wave asymmetry for the Fe bilayer on W(110) for wave vectors ranging from $-\bar{H}$ to \bar{H} in the two dimensional Brillouin zone, using theoretically determined Dzyaloshinskii-Moriya interaction parameters. The simulations were performed at room temperature, as in experiment, with realistic damping. We obtain a qualitatively good agreement with experiment but

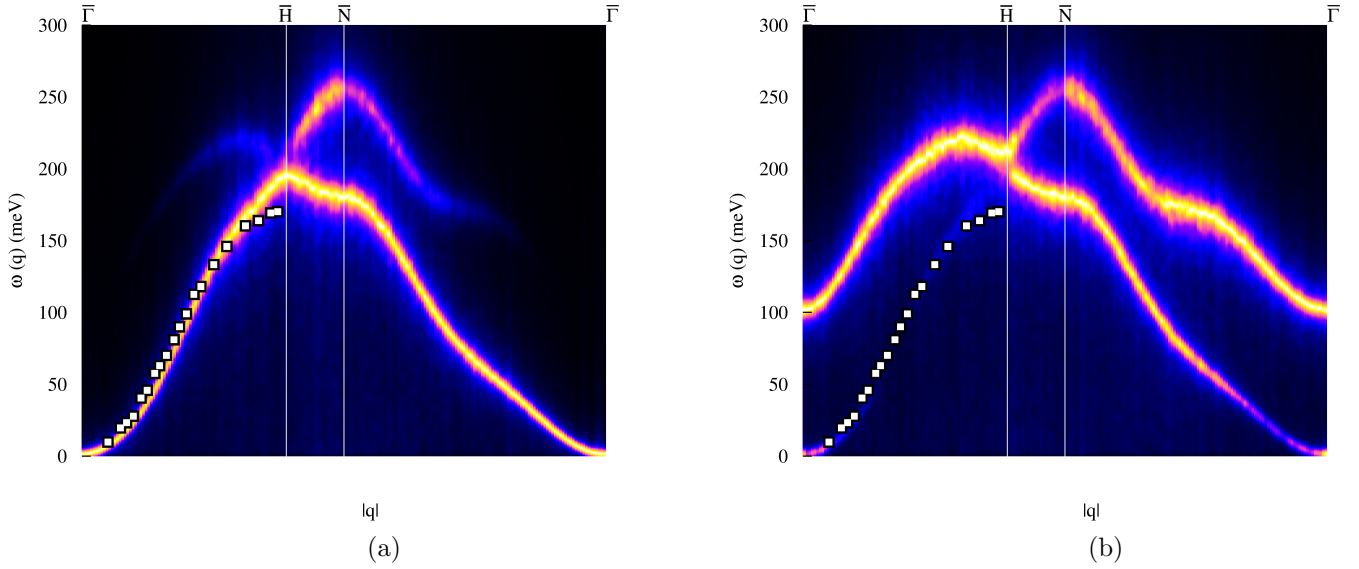


Figure 7. Spin wave dispersion spectra obtained from ASD simulations of 2 ML Fe/W(110) at $T=300$ K and $\alpha = 0.01$. a) Sampling inside the first Brillouin zone ($\boldsymbol{\tau} = [00]$) b) Sampling shifted by vector $\boldsymbol{\tau} = [10]$. Experimental values are marked by white squares.⁵¹ See the text for details.

the amplitude of the calculated spin wave asymmetry is slightly overestimated ($\approx 12\text{meV}$ compared to $\approx 8\text{meV}$ in experiment). There are several explanations to this discrepancy, primarily the asymmetry is sensitive to the value of the Dzyaloshinskii-Moriya interaction which is very delicate to calculate from ab-initio theory. In particular, it requires many k-points in order to converge as well depends a bit on technical parameters such as basis-set, sphere radii and choice of exchange correlation functional. The assumption made in the calculations of an atomically sharp interface between Fe and W may also be a limiting factor.

C. Analysis of the dynamical structure factor

In neutron scattering, the susceptibility can be written in the following form⁵³

$$\bar{\chi}(\mathbf{q} + \boldsymbol{\tau}, \omega) = \frac{1}{2}(1 + \cos\phi)\bar{\chi}_{Ac}(\mathbf{q}, \omega) + \frac{1}{2}(1 - \cos\phi)\bar{\chi}_{Op}(\mathbf{q}, \omega), \quad (9)$$

where $\bar{\chi}_{Ac}$ and $\bar{\chi}_{Op}$ are the susceptibilities originating from acoustic and optical branch, respectively, \mathbf{q} is a reciprocal vector within the primitive Brillouin zone (BZ), $\boldsymbol{\tau} = [hkl] = h\mathbf{b}_1 + k\mathbf{b}_2 + l\mathbf{b}_3$, $\phi = \boldsymbol{\tau} \cdot \boldsymbol{\rho}$, where $\boldsymbol{\rho}$ is a vector connecting two sublattices. In this manner,

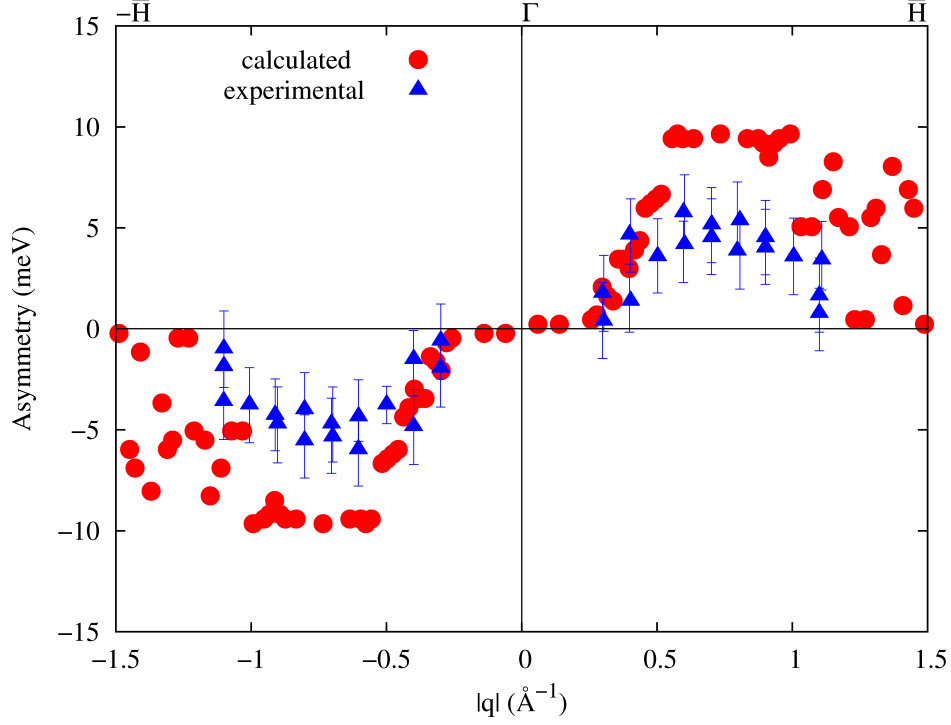


Figure 8. (Color online) Calculated spin wave asymmetry for the magnon spectrum of 2 ML Fe/W(110), using theoretically determined Dzyaloshinskii-Moriya interactions. The experimental values have been obtained by Zakeri et al.⁴ for $\mathbf{M} \parallel [\bar{1}10]$.

by changing the momentum transfer by varying $\boldsymbol{\tau}$, the intensity of the acoustic and optical branches is changing. If we take the Fe bilayer on W(110) as an example, $\boldsymbol{\tau} = [0 \frac{1}{\sqrt{2}} 0.5]a$ and the reciprocal vectors are restricted in the film-plane ($l=0$), then it follows that $\phi = h\pi + k\pi$. Inside the primitive BZ, the phase $\phi = 0$ and in the limit $\mathbf{q} \rightarrow \mathbf{0}$, in Eq. (9) the acoustic term will dominate and will be detected in experiment. If we go outside the first BZ, it is possible to have a situation where the optical term dominates, on the expense of the acoustic response, for instance by choosing $\boldsymbol{\tau} = [10]$, as illustrated in Fig. 7(b). If there are more than two atoms in the unit cell, the analysis becomes more complicated but the principle is the same.

III. CONCLUSIONS

Summarizing, we have shown that combining first principles calculations with atomistic spin dynamics simulations provides a powerful tool for studies of magnetic excitations in low dimensional systems. Our method can be applied to other low-dimensional systems,

particularly for cases in which exotic non-collinear ground states occur.^{5,9,10} It is hoped that the remarkable progress using the SPEELS technique³ will continue to provide new and surprising experimental results, for which the currently presented theory seems to be a good tool for analyzing the experimental data and predicting magnon spectra and related properties. The materials presented here have several common features, e.g. the absence of optical modes in the magnon curves, both as determined by experiments as well as obtained by theory. This fact has been analyzed in detail and it is argued that also the optical modes should be visible if one considers excitations which allow for momentum transfer outside the first Brillouin zone. The realization of this in the SPEELS method is clearly a challenge. In addition, we show that all thin film systems investigated here have a spin wave stiffness which is considerably softer compared to the bulk value. This applies both to Fe as well as Co films. Finally, we report on a quantitative agreement between theory and measured magnon curves for 2 ML Fe on W(110) and 8 ML Co on Cu(001).

ACKNOWLEDGMENTS

We gratefully acknowledge the European Research Council (ERC project 247062 - ASD), the Swedish Research Council (VR), Knut and Alice Wallenberg Foundation, Carl Tryggers Foundation and Göran Gustafsson Foundation for financial support. L.B. and A.B. also acknowledge SeRC and eSENCE, respectively. The computer simulations were performed on resources provided by the Swedish National Infrastructure for Computing (SNIC) at National Supercomputer Centre (NSC), UppMAX and High Performance Computing Center North (HPC2N). We thank B. Hjörvarsson, J. Kirschner, L. Nordström, E. Papaioannou, L. Szunyogh, L. Udvardi, D. Mills and Kh. Zakeri for valuable discussions.

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¹ M. Plihal, D. L. Mills, and J. Kirschner, Phys. Rev. Lett. **82**, 2579 (1999).

² R. Vollmer, M. Etzkorn, P. S. Anil-Kumar, H. Ibach, and J. Kirschner, Phys. Rev. Lett. **91**, 147201 (2003).

³ J. Prokop, W. X. Tang, Y. Zhang, I. Tudosa, T. R. F. Peixoto, Kh. Zakeri, and J. Kirschner, Phys. Rev. Lett. **102**, 177206 (2009).

- ⁴ Kh. Zakeri, Y. Zhang, J. Prokop, T.-H. Chuang, N. Sakr, W. X. Tang, and J. Kirschner, Phys. Rev. Lett. **104**, 137203 (2010).
- ⁵ L. Udvardi and L. Szunyogh, Phys. Rev. Lett. **102**, 207204 (2009).
- ⁶ L. Udvardi, L. Szunyogh, L. Palotás, and P. Weinberger, Phys. Rev. B **68**, 104436 (2003).
- ⁷ A. T. Costa, R. B. Muniz, J. X. Cao, R. Q. Wu, and D. L. Mills, Phys. Rev. B **78**, 054439 (2008).
- ⁸ A. Bergman, A. Taroni, L. Bergqvist, J. Hellsvik, B. Hjörvarsson, and O. Eriksson, Phys. Rev. B **81**, 144416 (2010).
- ⁹ M. Bode, M. Heide, K. von Bergmann, P. Ferriani, S. Heinze, G. Bihlmayer, A. Kubetzka, O. Pietzch, S. Blugel, and R. Wiesendanger, Nature (London) **447**, 190 (2007).
- ¹⁰ M. Heide, G. Bihlmayer, and S. Blügel, Phys. Rev. B **78**, 140403 (2008).
- ¹¹ A. T. Costa, R. B. Muniz, and D. L. Mills, Phys. Rev. B **74**, 214403 (2006).
- ¹² A. T. Costa, R. B. Muniz, and D. L. Mills, Phys. Rev. B **69**, 064413 (2004).
- ¹³ A. T. Costa, R. B. Muniz, and D. L. Mills, Phys. Rev. B **70**, 054406 (2004).
- ¹⁴ A. T. Costa, R. B. Muniz, S. Lounis, A. B. Klautau, and D. L. Mills, Phys. Rev. B **82**, 014428 (2010).
- ¹⁵ V. P. Antropov, M. I. Katsnelson, B. N. Harmon, M. van Schilfgaarde, and D. Kusnezov, Phys. Rev. B **54**, 1019 (1996).
- ¹⁶ B. Skubic, J. Hellsvik, L. Nordström, and O. Eriksson, J. Phys.: Condens. Matter **20**, 315203 (2008), <http://www.fysik.uu.se/cmt/asd/>.
- ¹⁷ “<http://www.physics.uu.se/en/page/uppasd>,” .
- ¹⁸ J. H. Mentink, M. V. Tretyakov, A. Fasolino, M. I. Katsnelson, and Th. Rasing, J. Phys.: Condens. Matter **22**, 176001 (2010).
- ¹⁹ I. A. Dzyaloshinskii, Sov. Phys. JETP **5**, 1259 (1957).
- ²⁰ T. Moriya, Phys. Rev. **120**, 91 (1960).
- ²¹ G. Kresse and J. Furthmüller, Comput. Mater. Sci. **6**, 15 (1996).
- ²² G. Kresse and D. Joubert, Phys. Rev. B **59**, 1758 (1999).
- ²³ H. Ebert, D. Ködderitzsch, and J. Minár, Rep. Prog. Phys. **74**, 096501 (2011).
- ²⁴ *The Munich SPR-KKR package*, version 5.4,
H. Ebert et al., Rep. Prog. Phys. **74**, 096501, 2011.
- ²⁵ H. Ebert and S. Mankovsky, Phys. Rev. B **79**, 045209 (2009).

- ²⁶ A. I. Liechtenstein, M. I. Katsnelson, and V. A. Gubanov, J. Phys. F: Metal Physics **14**, L125 (1984).
- ²⁷ A. Liechtenstein, M. Katsnelson, V. Antropov, and V. Gubanov, J. Magn. Magn. Mater. **54-57**, 965 (1986).
- ²⁸ J. Kübler, *Theory of Itinerant Electron Magnetism* (Oxford University Press, Oxford, 2009).
- ²⁹ X. Tao, D. P. Landau, T. C. Schulthess, and G. M. Stocks, Phys. Rev. Lett. **95**, 087207 (2005).
- ³⁰ K. Chen and D. P. Landau, Phys. Rev. B **49**, 3266 (1994).
- ³¹ S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Oxford University Press, Oxford, 1984).
- ³² D. P. Landau and K. Binder, *A Guide to Monte Carlo Simulations in Statistical Physics* (Cambridge University Press, Cambridge, 2000).
- ³³ C. A. F. Vaz, J. A. C. Bland, and G. Lauhoff, Rep. Prog. Phys. **71**, 056501 (2008).
- ³⁴ A. Taroni, S. Bramwell, and P. Holdsworth, J. Phys.: Condens. Matter **20**, 275233 (2008).
- ³⁵ E. Holmström, L. Nordström, L. Bergqvist, B. Skubic, B. Hjörvarsson, I. A. Abrikosov, P. Svedlindh, and O. Eriksson, *Proceedings of the National Academy of Sciences of the United States of America* **101**, 4742 (2004).
- ³⁶ M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Phys. Rev. Lett. **85**, 5424 (2000).
- ³⁷ N. M. Rosengard and B. Johansson, *Phys. Rev. B* **55**, 14975 (1997).
- ³⁸ S. V. Halilov, H. Eschrig, A. Y. Perlov, and P. M. Oppeneer, Phys. Rev. B **58**, 293 (1998).
- ³⁹ M. Pajda, J. Kudrnovský, I. Turek, V. Drchal, and P. Bruno, Phys. Rev. B **64**, 174402 (2001).
- ⁴⁰ F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, *Physical Review B* **49**, 3962 (1994).
- ⁴¹ H. J. Elmers, J. Hauschild, H. Fritzsche, G. Liu, and U. Gradmann, Phys. Rev. B **75**, 2031 (1995).
- ⁴² U. Gradmann, “Handbook of magnetic materials,” (Elsevier, Amsterdam, 1993) Chap. 1.
- ⁴³ R. N. Sinclair and B. N. Brockhouse, Phys. Rev. **120**, 1638 (1960).
- ⁴⁴ M. Etzkorn, P. S. Anil-Kumar, R. Vollmer, H. Ibach, and J. Kirschner, Surface Science **566-568**, 241 (2004).
- ⁴⁵ R. Vollmer, M. Etzkorn, P. S. Anil-Kumar, H. Ibach, and J. Kirschner, J. Appl. Phys. **95**, 7435 (2004).
- ⁴⁶ P. Mohn, *Magnetism in the Solid State* (Springer, Berlin, 2003).
- ⁴⁷ P.-W. Ma and S. Dudarev, Phys. Rev. B **86**, 054416 (2012).

- ⁴⁸ R. Chimata, A. Bergman, L. Bergqvist, B. Sanyal, and O. Eriksson, Phys. Rev. Lett. **109**, 157201 (2012).
- ⁴⁹ A. Taroni, A. Bergman, L. Bergqvist, J. Hellsvik, and O. Eriksson, Phys. Rev. Lett. **107**, 037202 (2011).
- ⁵⁰ L. M. Sandratskii, Phys. Rev. B **81**, 064417 (2009).
- ⁵¹ W. X. Tang, Y. Zhang, I. Tudosă, J. Prokop, M. Etzkorn, and J. Kirschner, Phys. Rev. Lett. **99**, 087202 (2007).
- ⁵² Kh. Zakeri, Y. Zhang, T.-H. Chuang, and J. Kirschner, Phys. Rev. Lett. **108**, 197205 (2012).
- ⁵³ J. Jensen and A. Mackintosh, *Rare Earth Magnetism* (Oxford University Press, Oxford, 1991).